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Progress Reports

MOUND LABORATORY PROGRESS REPORT FOR JULY, 1963

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The Mound Laboratory Progress Report, issued monthly, is intended to be a means of reporting items of current technical interest in research and development programs. To issue this report as soon as possible after the end of the month, editorial work is limited; and since this is an informal progress report, the results and data presented are preliminary and subject to change.

These reports are not intended to constitute publication in any sense of the word. Final results either will be submitted for publication in regular professional journals or will be published in the form of MLM topical reports.

The previous reports in this series are:

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MONSANTO RESEARCH CORPORATION

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DISCLAIMER

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SUMMARY

Radioelements

Thallium-204 Half-life Thallium-204 was tentatively assigned a half-life of 3.781 ± 0.010 years based on calorimetric measurements. The calorimetric assay of the sample will be continued for several years to decrease the probable error.

Polonium-208-Polonium-209 Decay Scheme Studies The decay scheme for polonium-208-polonium-209 isotopic mixtures is being determined. The gamma spectrum is being studied using two sources of different isotopic composition. Definite peaks have been established at 76, 260, 300, 575, and 910 kev. Various multiple coincidence experiments indicated that the 300-and 575-kev photons and the 76-kev x-ray are in triple coincidence. Relative intensities of the various gamma rays with the two different sources indicated that the 575-kev photon is associated with polonium-208 decay, while the 260- and 910-kev states are attributed to polonium-209.

A theoretical estimate of the partial alpha half-life for polonium-208 decay to the 2+ level in lead-204 (at 0.899 Mev) has been made. This value (2.5×10^6 years) was determined graphically by extrapolation to an alpha particle energy of 4.211 Mev. Polonium-208-polonium-209 sources are being prepared for the investigation of the internal conversion electron spectra of these isotopes.

Actinium-227 Separation Ion exchange techniques are being studied for the separation of actinium-227 from thorium-227, radium-223, and radium-226. Naturally occurring lanthanum, thorium, and barium are being used as experimental substitutes. Composite elution curves indicate that lanthanum and thorium-232 can be successfully isolated with two molar hydrochloric acid, three molar nitric acid, and ammonium acetate as elutrients. A purified form of the original Dowex 50W-X8 column packing material was used; the new material (AG-50W-X8) reduced tailing on the elution curves.

Thorium-228 Half-life Thorium-228 was tentatively assigned a half-life of 1.9132 years with an internal probable error of 0.0005 year. The presence of polonium-210 and actinium must be positively identified in the sample by chemical analysis and alpha pulse height analyses.

Uranium-234 Separation and Analysis Mound Laboratory is studying the separation and purification of uranium-234 from aged plutonium-238 using ion exchange and solvent extraction techniques. A batch containing en estimated 250 milligrams of uranium-234 in 26.2 grams of plutonium is being processed. The anion exchange separation procedure for this batch was completed. More than 99 per cent of the plutonium was removed from the uranium.

A spectrophotometric method is being developed for the determination of uranium-234 in samples containing aluminum, iron, and plutonium. The extraneous metallic ions were separated, and the uranium was complexed with 1-(2-pyridylazo)-2-naphthol (PAN). After the color developed, the absorbance was read at 570 millimicrons. Standard uranium samples were analyzed, and Beer's law was followed between 0 and 80 micrograms of uranium.

Isotope Separation

Carbon-13 Gram quantities of over 90 per cent pure carbon-13 are being prepared. Various thermal diffusion column configurations are being used for separation efficiency studies and evaluation of feed gases, such as carbon monoxide or methane.

The first five stages of the seven-stage cascade system of hot-wire and concentric-tube columns are enriching natural methane to about 23 per cent carbon-13 while coming to steady state conditions. The concentric tube columns enriched methane from 60 per cent to over 90 per cent carbon-13 in the total carbon. About 1.5 grams of product containing about 10 per cent impurities have been accumulated.

Thermal Diffusion Column Wire Heating by Alternating Current The use of alternating current for heating thermal diffusion column wires is being studied. Vibration of the wire is the major problem. Two parallel iron discs were used to align the electrical leads and the column wire in the pool of mercury; however, the discs did not prevent vibration. The mercury pool was replaced by rubber bands, and subsequently by a spring, for tensioning the wire; there was a slight vibration. A braided cable was used as an electrical lead for alternating current and as a vibration damper. Both the vibration and the use of a mercury pool were eliminated.

Alpha and Neutron Source Development

Neutron Measurements An integral has been set up to determine the error introduced into the calibration of neutron sources by the finite geometry of a source. For a source with 80 grams of plutonium, the effect of geometry gave a calibration 0.25 per cent higher than for a point source.

The ratio of room-scattered neutrons to unscattered neutrons in a counting room is being determined. An empirical expression for the ratio as a function of source-to-detector separation has been revised. In the revised expression there are only two parameters as compared to three in the earlier form.

Polonium Alpha Sources Several different polonium alpha sources were prepared by different plating techniques. As base material copper received a more uniform coating than platinum. A silver strike bath on the polonium allowed a thinner, wipe-free gold plating. Rouge has been substituted for scouring powder, which cut grooves in a gold plate during swaging of the plate.

Analytical

Low-level Krypton Counting Experiments with a mixture of xenon containing 1.3×10^{-9} atom per cent krypton-85 and 10 per cent methane indicated that the counting chamber efficiency is 68 per cent and that the counter will detect 2.9×10^{-13} atom per cent krypton-85.

Four Pi Beta Counting A study was made to verify that the counts in coincidence between the two hemispheres of the four pi beta counter are the results of cross scattering. The number of coincidences increased with the aperture size; however, the increase was not strictly linear with aperture area due to such factors as counting statistics, instrument resolving time uncertainty, and penetration of the lead holder by scattered particles. The data show that the preponderant effect of coincidence is due to cross scattering.

RADIOELEMENTS RESEARCH

Basic and applied research on a number of radioelements is being conducted to determine their physical properties, develop analytical techniques, and study the basic radiochemistry involved. Of particular interest are alpha emitters, their decay chains, their isotopes, and their chemical homologs.

Half-life of Thallium-204

A neutron-irradiated thallium source has been assayed periodically in Calorimeter 58 since February 1, 1958. At the end of 35 days the effective half-life was reported as 2.76 years. The possibility that this half-life value was low due to the presence of a shorter lived isotope was later confirmed. An activity with an approximately eighteen-day half-life has been found in the least squares solution determined with early data points. Probably the isotope was formed in the inner aluminum jacket containing the thallium. The outer jacket was removed after irradiation and replaced with one of the same type.

An analysis of the data obtained between 211 and 1162 days indicated that no other activity with a shorter half-life than thallium was present. The half-life determined for thallium-204 in this period was 3.781 ± 0.010 years (absolute probable error). The data were not solved for activities with longer half-lives than thallium; however, the half-life of the sample from 211 to 779 days was 3.803 ± 0.036 years, indicating no significant trend of the half-life as a function of time. A gamma pulse height analysis of the sample (October 16, 1962) showed weak gamma peaks at 440, 900, 1220, and 1390 kev. Since thallium-204 is believed to have no gamma activity, there may be other isotopes present that have not significantly affected the calorimeter results. Measurements will be continued, and results will be reported yearly.

Polonium-208-Polonium-209 Decay Scheme Studies

The decay schemes for samples of polonium-208-polonium-209 on hand at Mound Laboratory are being determined. For reference a single spectrum of a polonium-208-polonium-209 source is shown in Figure 1. Coincidence spectra were obtained with matched three-inch by three-inch NAI(Tl) detectors and by gating separately on all the main peaks through the fast-slow coincidence circuit. Double delay line amplifiers and zero crossover trailing edge pickoff are currently being used.

Results of the coincidence measurements are shown in Figures 2, 3, 4, and 5. Figure 2 shows that setting the gate on the x-ray peak yields a spectrum with photopeaks at 76, 300, 575, and 910 kev. No evidence exists here of a peak at 260 kev, as in the singles spectrum. The 300-, 575-, and 910-kev states are attributed to electron capture decay either in polonium-208 or polonium-209, that is, to excited states in bismuth-208 or bismuth-209. This conclusion is not in contradiction to unpublished values appearing in nuclear data tables.[1] A peak at 76 kev is probably due to triggering of the gate detector with scattered Compton gamma rays from 300-, 575-, and 910-kev states or from partial internal conversion of these states. The coincidence spectrum obtained by gating on the 575-kev peak is shown in Figure 3. The x-ray peak appears as expected; hence, through electron capture there exists a cascade of two gamma rays through transitions of 300 and 575 kev by this mode. A second peak appears at approximately 375 kev. Comparison of the two spectra shown in Figure 3, however, shows that at higher source-detector geometry, the 375-kev peak is more pronounced. This peak is the result of simultaneous detection of both the 300-kev and x-ray quanta, which yield a sum peak. The ratio of the 300- to 375-kev peaks will thus change with geometry and source intensity. The broadened peak at 140 kev is probably due to 180-degree scatter of the 300-kev photon. The peak at 220 kev in the high geometry measurement is attributed to summation of the 180-degree scatter radiation and the x-ray.

[1] D. Strominger, J. M. Hollander, and G. T. Seaborg, *Revs. Modern Phys.*, **30**, 585 (1958)

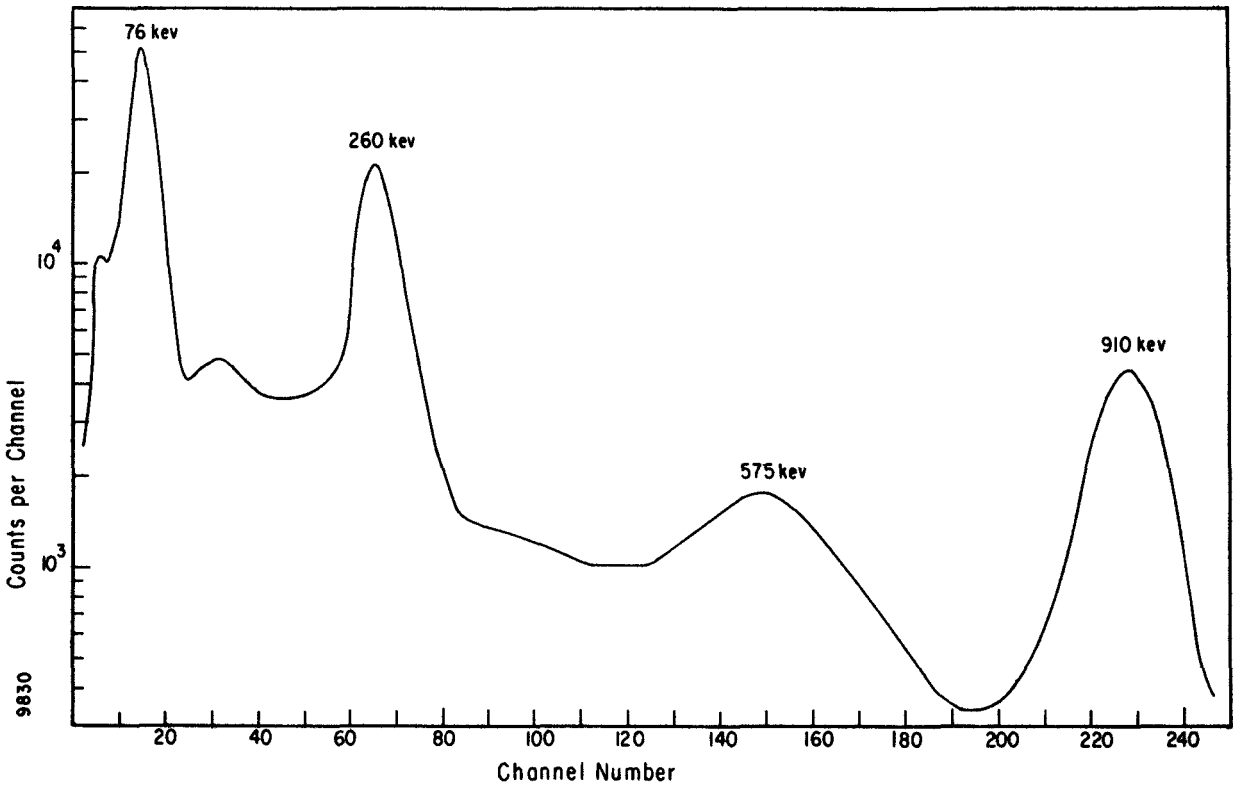

Figure 1. *Singles Spectrum of Polonium-208-Polonium-209 Source.*

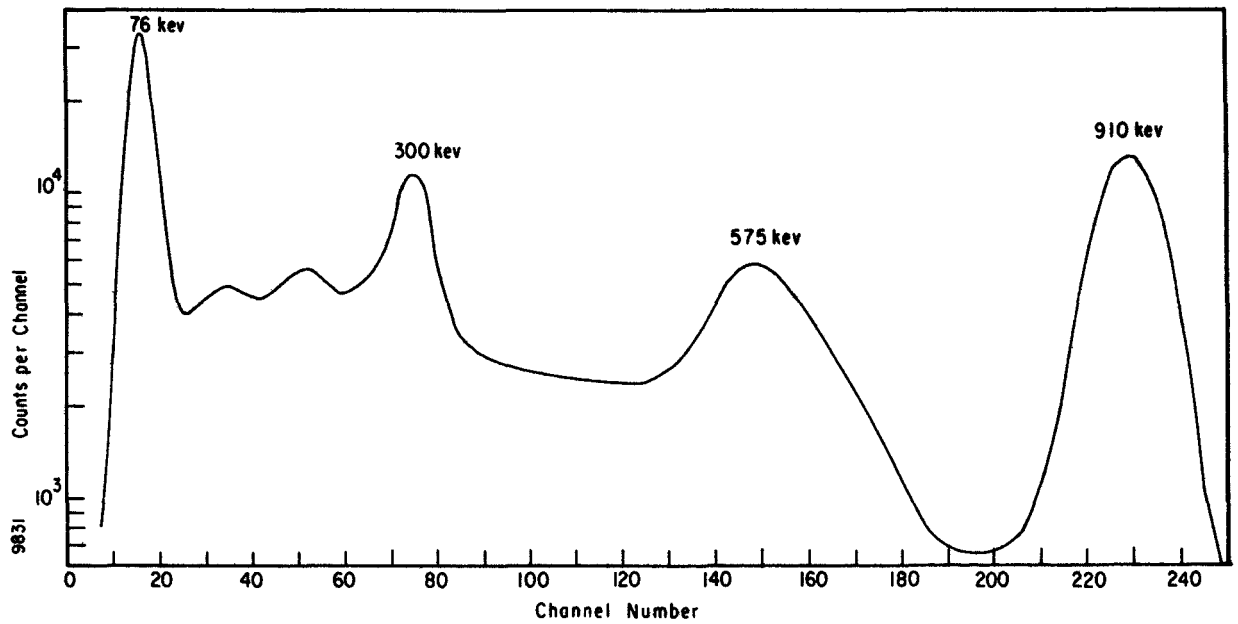

Figure 2. *Coincidence Spectrum of Polonium-208-Polonium-209: 20-kev gate on x-ray (76 kev) peak; 925-minute measurement; resolving time 75 nanoseconds.*

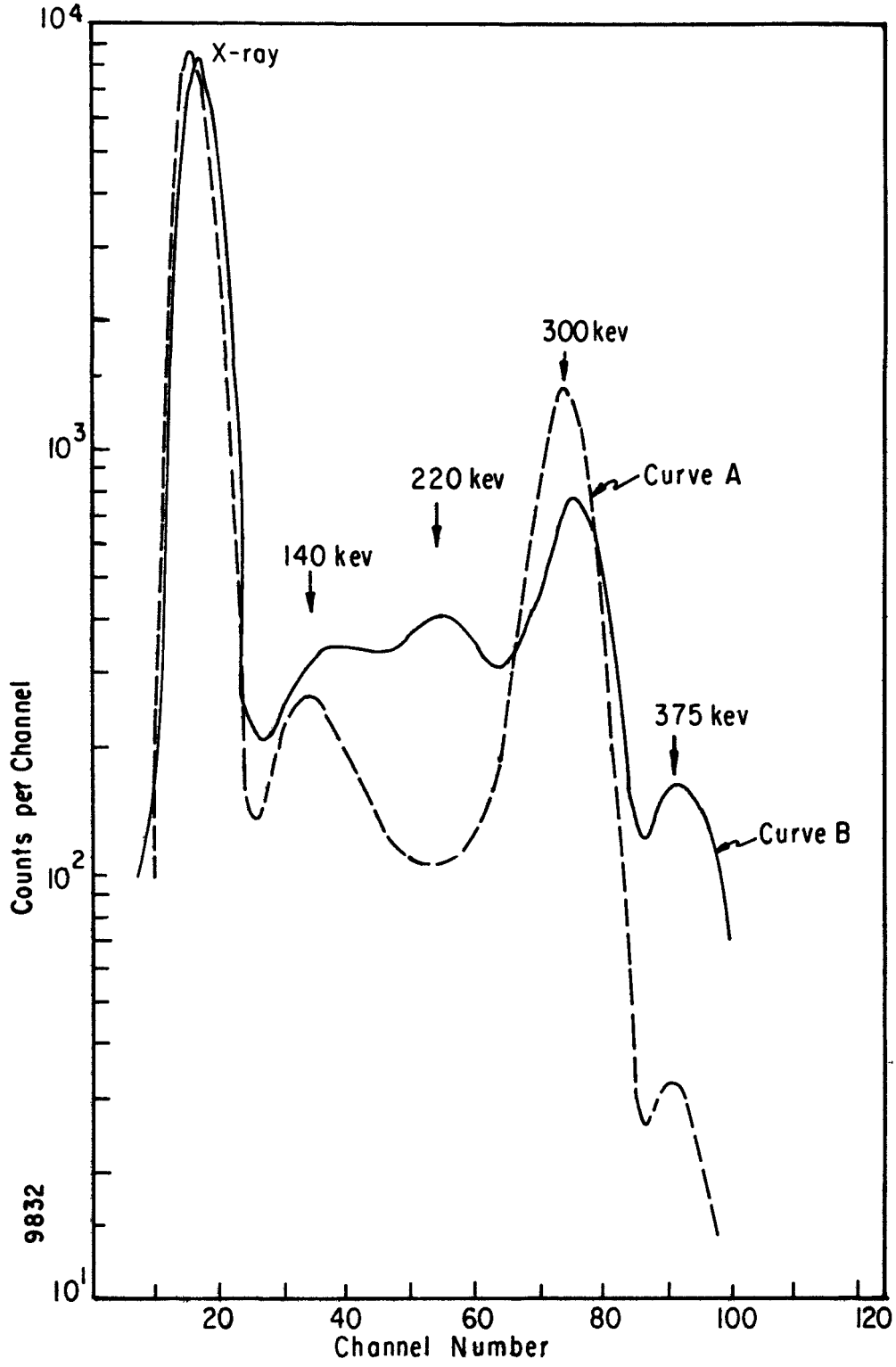

Figure 3. *Coincidence Spectrum of Polonium-208-Polonium-209: 50-kev gate set on 575-kev peak. Curve A: low source-detector geometry; 1000-minute measurement. Curve B: high source-detector geometry; 200-minute measurement; resolving time 75 nanoseconds.*

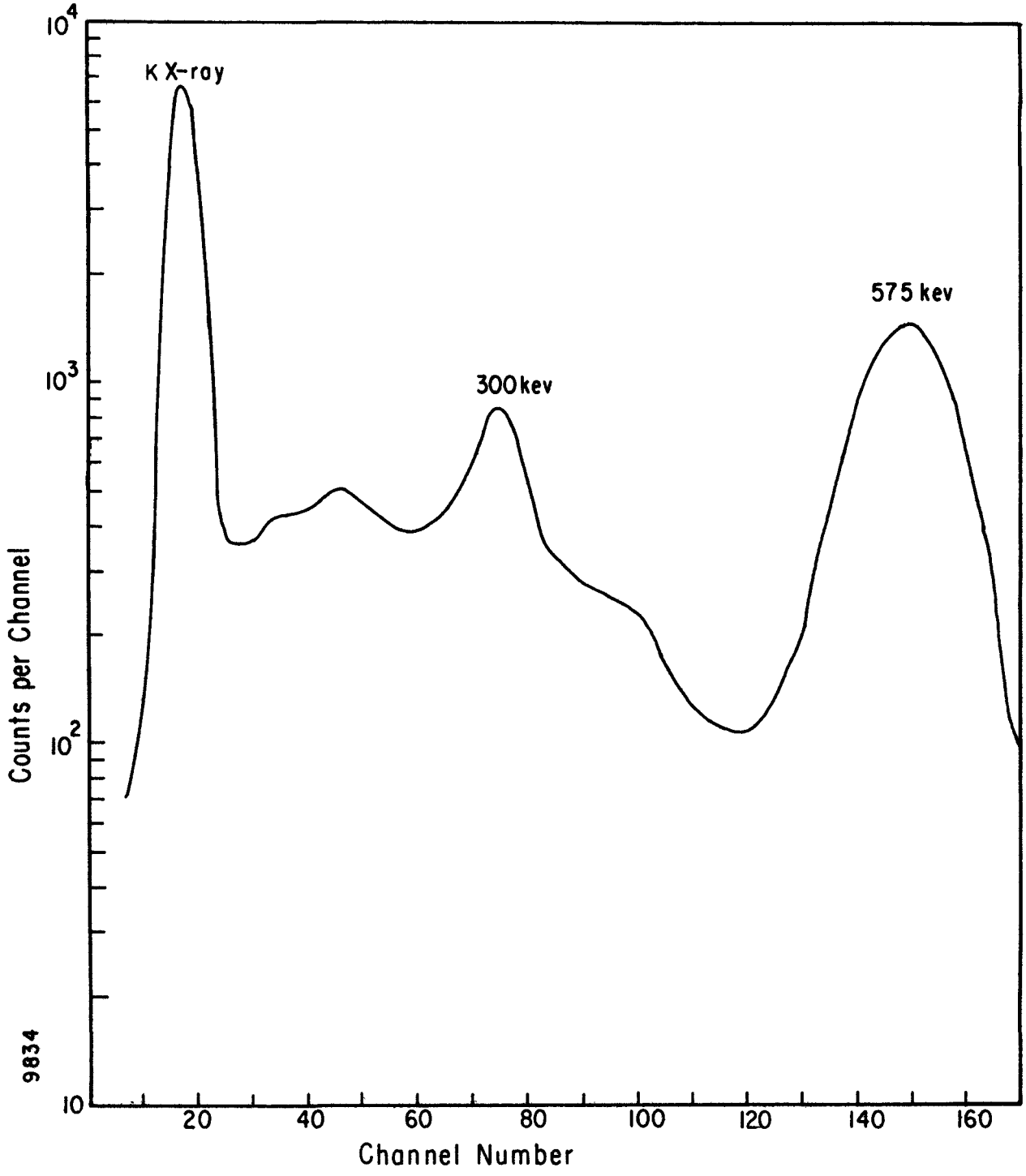

Figure 4. *Coincidence Spectrum of Polonium-208-Polonium-209: 20-kev gate at 300 kev; resolving time approximately 75 nanoseconds; 1000-minute measurement.*

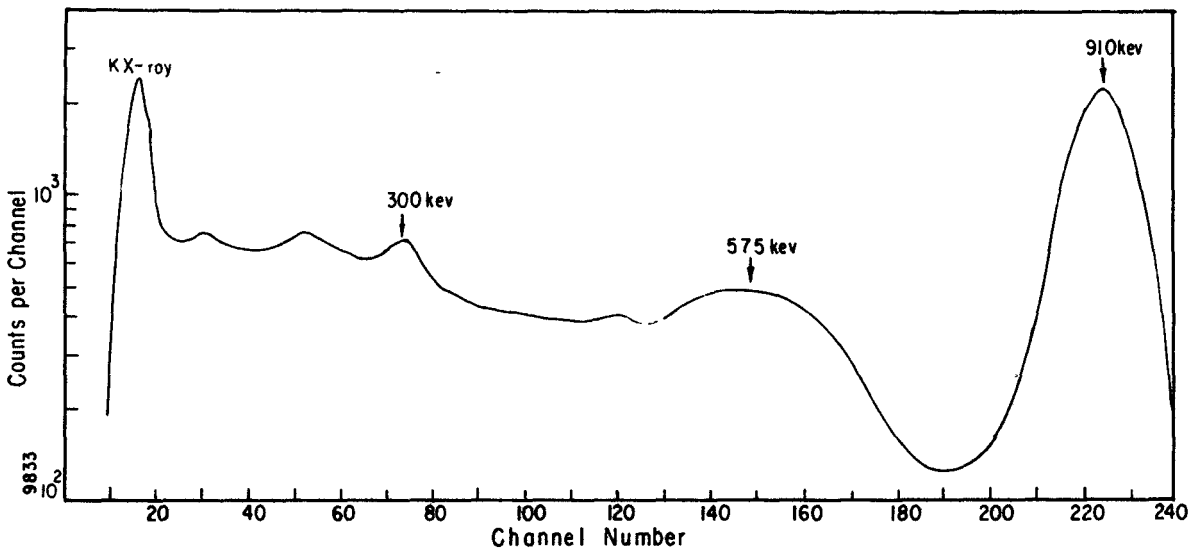

Figure 5. *Coincidence Spectrum of Polonium-208-Polonium-209, Source DIS-67: 30-kev gate on x-ray peak; 3780-minute measurement; resolving time 75 nanoseconds.*

The result of gating on the 300-kev peak is shown in Figure 4. The 575- and 76-kev peaks are expected, whereas the 300-kev state shows up as a result of triggering of the gate detector by scattered Compton quanta from the 575-kev state. Selecting gate conditions, even below 300 kev down into the region of the 260-kev state, yields essentially the same results. From these observations the 260-kev gamma ray does not appear to be related to any of the other gamma transitions. It has tentatively been associated with alpha decay although this association must be substantiated. The 575-kev peak in this spectrum is broad and shows evidence of a state on the upper side at approximately 640 kev.

Further investigations indicate a multiplicity of states in the 575-kev region; however, these data are being analyzed further. The 910-kev state has been shown to be in coincidence with the x-ray only; the spectrum exhibiting this coincidence is not shown.

Time delays have been imposed in addition to amplitude restrictions in coincidence studies. Preliminary results show that earlier indications of delayed states were caused by a time jitter in the RC-clipped amplifiers using leading edge triggering. Spectral distortion also results in trailing edge operation, but it is less severe if the time gate is not reduced below 60 nanoseconds. Possible delayed states will be investigated further.

Some preliminary analyses of singles spectra have been made by spectral stripping, that is successively peeling off the various constituent gamma rays. This is being done by obtaining the spectrum of monoenergetic sources similar in energy to the pertinent gamma rays in the polonium-208-209 spectrum. These spectra are then adjusted for differences in resolution and peak-to-Compton ratio and adjusted to the polonium-208-209 spectrum. For example, the manganese-54 spectrum (E_γ = 0.84 Mev) is being used to derive the total contribution of the 910-kev state. The 575-kev state cannot be fitted with a single gamma ray of this energy since the peak is broadened. The contributions to this peak may be obtained by fitting with two gamma rays of slightly different energies. Appropriate nuclides in this energy range are presently being sought.

Polonium-208-polonium-209 source material is available at Mound with different ratios of the two isotopes. By the use of this material various states will be assigned to either polonium-208 or polonium-209. Varying isotopic compositions have not yet been used for investigation of these nuclides. The following conclusions are tentative since experimental data have not been completely analyzed. In addition further experiments are being performed in this investigation.

Singles spectra of two polonium-208-209 sources were presented previously.[2] Source *DIS*-67 had a substantially smaller quantity of polonium-208, and the 575-kev gamma ray was absent; whereas, the 260-kev and 910-kev states were in about the same ratio in both sources. The 575-kev state has thus been associated with polonium-208 decay, whereas, the 260- and 910-kev states are attributed to polonium-209 decay. X-radiation arises from a number of processes, and it is associated with both nuclides.

The results of coincidence measurements on *DIS*-67, with a 30-kev gate around the x-ray peak, is shown in Figure 5 and should be compared with Figure 2 discussed above. Source *DIS*-67 shows a predominant 910-kev peak and slight evidence of states at 575 and 300 kev. These states are to be expected since the 575-kev state is associated with polonium-208 decay. Specifically, the 300- and 575-kev cascades, which are coincident with the x-ray, are associated with electron capture decay in polonium-208. Predominance of the 910-kev state in this coincidence spectrum is further evidence for its assignment to polonium-209 since it is also in coincidence with the x-ray.

A theoretical estimate of the partial alpha half-life $[(t_{1/2})_\alpha]$ for polonium-208 decay to the 2+ level in lead-204 (at 0.899 Mev) has been made. The partial half-life for alpha-particle transition to a 2+ state in the daughter nucleus is not seriously hindered for even-even nuclei. Hindrance factors at 1.5 to 2.0 are common. The $(t_{1/2})_{\alpha 899}$ for the polonium-208 to lead-204 transition can be estimated by extrapolation of a graph of $(t_{1/2})_\alpha$ as a function of alpha particle energy. Since there is a sharp drop in alpha binding energy at shell closure at neutron number N = 126, the graph must be constructed for polonium isotopes with $N \leq 126$. Figure 6 was constructed from literature values[3] for $(t_{1/2})_\alpha$ and alpha particle energy. Extrapolation to an alpha particle energy of 4.211 Mev leads to a partial alpha half-life of 2.6×10^6 years. The ratio of $(t_{1/2})_{\alpha 899}$ to $(t_{1/2})_{\alpha 0}$ is then 1.1×10^{-5}. Neglecting internal conversion of the 0.899-Mev gamma ray, 1.0 millicurie of polonium-208 would emit 2.4×10^3 gammas per minute at 0.899 Mev. A search for a gamma ray of this energy in coincidence with an alpha particle in the decay of polonium-208 is planned.

A source of approximately 30 microcuries of polonium-208-polonium-209 was prepared for an investigation of the internal conversion electron spectra of these isotopes. The polonium was evaporated on Mylar-backed aluminum to minimize backscatter. Some contamination of the semiconductor electron detector and the detector vacuum chamber occurred when the source was in use; nevertheless, usable data were obtained. A source is now being prepared on Mylar-backed silver; with this source contamination is expected to be less of a problem, since the polonium will be electroplated onto the silver.

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[2] MLM-1155.
[3] *Ibid.*

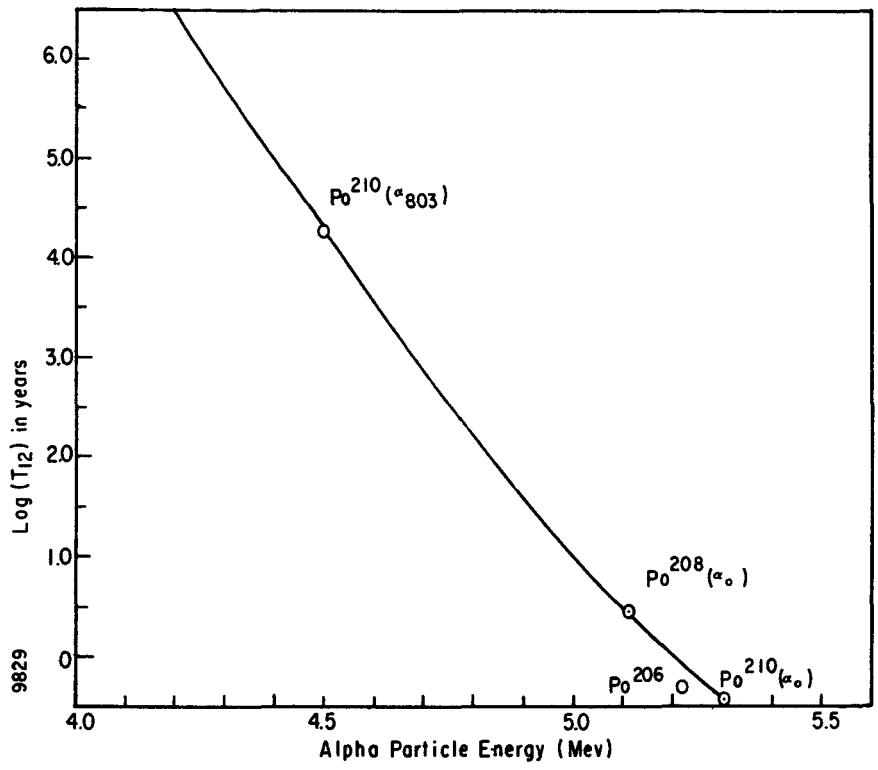


Figure 6. *Partial Alpha Half-life of Even-Even Polonium Isotopes as a Function of Alpha Particle Energy.*

Actinium-227 Separation

Ion exchange techniques are being studied for the separation of actinium-227 from thorium-227, radium-223, and radium-226. To develop the technique, naturally occurring isotopes were substituted for the highly radioactive materials (Table 1).

Table 1

EXPERIMENTAL SUBSTITUTES FOR ACTINIUM-227 SEPARATION[a]

Radioactive Ion	Substitute
Actinium-227	Lanthanum
Thorium-227	Thorium-232
Radium-223, 226	Barium
Lead-207, 211	Lead
Bismuth-211	Bismuth

[a]All sample solutions contain one mg metal per ml of 2*M* HCl.

Barium and lanthanum can be separated with Dowex 50W-X8 using two molar hydrochloric acid, three molar nitric acid, and six molar nitric acid as elutrients. However, thorium remains on the column under these conditions.

Ammonium acetate eluted thorium effectively from AG-50W-X8 (purified Dowex 50W-X8) as shown in Figure 7b. Test results (Figures 7a and 7c) also indicate that thorium and lanthanum can be separated with this column material. The elution characteristics of lead, bismuth, lanthanum, and barium from AG-50W-X8 will be studied further since less tailing was observed when this purified resin was used.

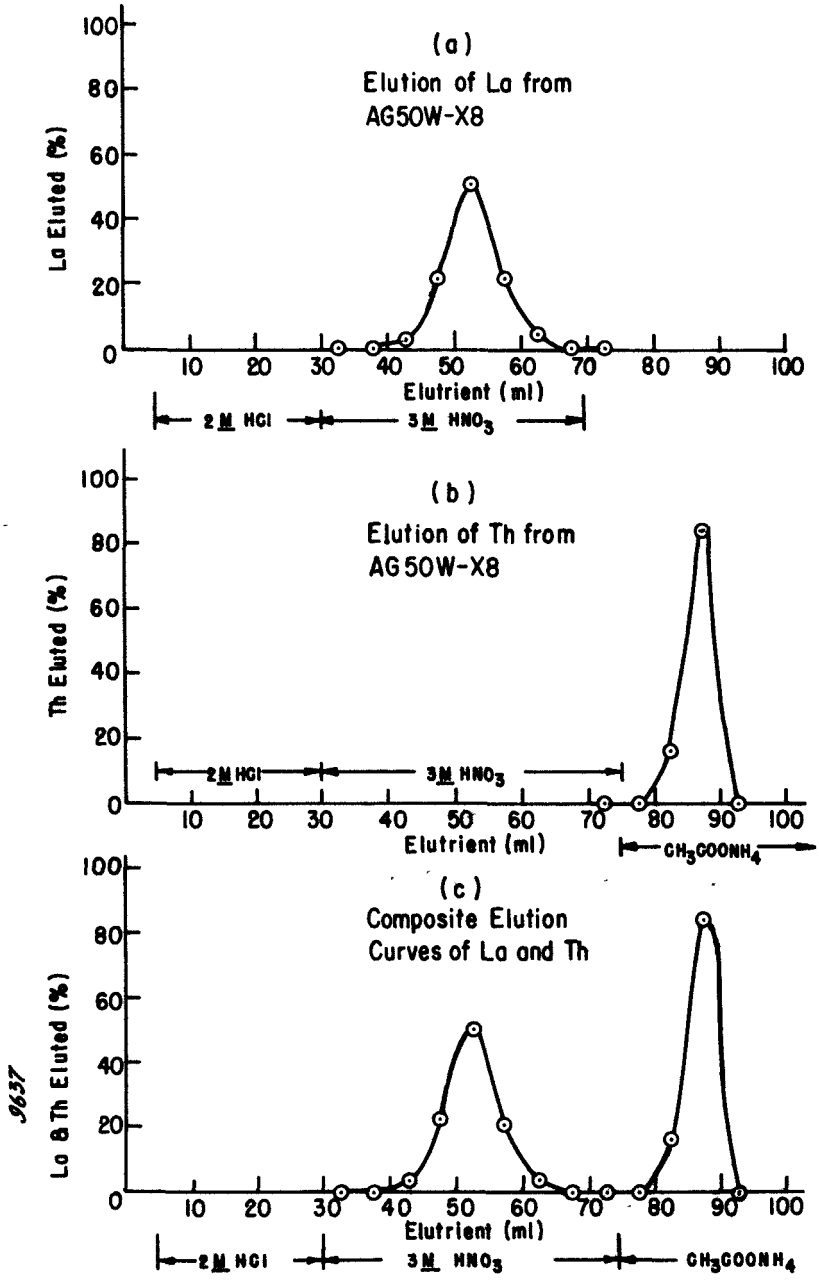


Figure 7. *Elution Curves for the Isolation of Lanthanum and Thorium.*

Thorium-228 Half-life

A thorium-228 sample (designated T-2) was obtained from the Cave Recovery Project of 1954 in which actinium-227 and thorium-228 were recovered from neutron-irradiated radium-226. The first five measurements on the sample (3 to 27 days after its separation on November 30, 1954) were made in Calorimeter 62, located in the Cave area. The measurements were continued, starting at 87 days, in Calorimeter 58.

The early data up to 295 days have not been analyzed completely. At the time of separation the sample probably contained the following isotopes and approximate curie contents:

(1) 0.5473 curies of thorium-228, which has 3.64-day radium-224 as the longest half-life product in the decay chain.

(2) 0.078 curies of 18.73-day thorium-227 with 11.44-day radium-223 being the longest half-life in the decay chain.

(3) 0.033 curies of polonium-210.

(4) 0.00089 curies of actinium-227 with 18.73-day thorium-227 and 11.44-day radium-223 in the decay chain.

The thorium-227 in Item (2) was originally the daughter of actinium-227 in the neutron-irradiated radium-226 slug, and the polonium-210 in Item (3) was from the radium-226 chain. Item (4) may be radium-226 instead of actinium-227, in which case there would be 0.00084 curies of radium-226 present.

Since there are at least six half-lives represented in the four items above, it is not possible to solve for the half-life of thorium-228 using all of the data simultaneously. Therefore, the data were analyzed beginning at 295 days, at which time the three shorter half-lives would no longer influence the results. The results are presented in Table 2. Positive identification of polonium-210, actinium-227, or radium-226 would allow the determination of thorium-228 to an internal probable error of about 0.0005 years. Polonium-210 is identified to a great extent by the data, requiring a half-life between 138.38 days (Po^{210}) and 165 days. No better fit is obtained with actinium-227 than with radium-226; however, from other considerations, such as chemistry and gamma pulse height analyses, actinium-227 is the better isotope for this calculation. Column 1 of Table 2, therefore, gives the most probable results; the half-life of thorium-228 is 1.9132 years. This value compares well with the alpha-counted half-life of 1.910 ± 0.002 years as reported by H. W. Kirby et al.[4] The sample used in Kirby's study was produced by the same nuclear reactions as the one considered in this study.

[4] H. W. Kirby et al., *Phys. Rev.*, **102**, No. 4, 1140-41 (1956).

Table 2

VARIATIONAL LEAST SQUARES SOLUTION OF THORIUM-228 HALF-LIFE DATA, ASSUMING THREE SPECIES PRESENT: A(Th^{228}), B(Ac^{227} or Ra^{226}), and C(Po^{210} or unknown)[a]

	Half-life ($T_{1/2}$)			
	B = 21.77 yrs C = 138.38 days	B = 1620 yrs C = 138.38 days	B = 21.77 yrs C = 165 days	B = 1620 C = 165
Most Probable Half-life of A (yrs)	1.91319	1.91443	1.91463	1.91584
Minimum Sum of the Deviations Squared $(\mu w)^2$	2364.2	2397.8	2367.6	2367.6
Probable Error of Half-life of A (yrs)	0.00050	0.00046	0.00057	0.00053
Probable Errors per Single Observation	5.99	6.03	5.99	5.99
Most Probable Power of A (μw)	110,396.0	110,431.2	110,312.2	110,342.5
Most Probable Power of B (μw)	179.5	127.0	168.2	118.0
Most Probable Power of C (μw)	1066.5	1100.5	1018.5	1052.0

[a]The equation for the determination of sample power as a function of time [w(t)] is:

$$W(t) = A \exp[\lambda(A)t] + B \exp[-\lambda(B)t] + C \exp[-\lambda(C)t]$$

where 34 data points extending from t = 295 to 3104 days were used; t = 0 was November 30, 1954.

Uranium-234 Separation and Analysis

A technique to separate uranium-234 from aged plutonium-238 solutions has been developed. Uranium is separated from a nitrate solution by an anion exchange process followed by a hexone extraction of the solution, and a thenoyltrifluoroacetone (TTA) extraction to remove the last traces of plutonium solution.

One batch of feed solution has been processed through these three steps; fourteen milligrams of uranium-234 were recovered. A second batch (containing 26.2 grams total plutonium calculated on basis of 41.85 per cent plutonium-238) was processed through the anion exchange step, and the solution containing the uranium fraction is presently being evaporated prior to the hexone extraction. This batch contains an estimated 250 milligrams of uranium-234.

A spectrophotometric method is being developed for the determination of uranium-234 (the method is sensitive to two micrograms.) in sample solutions containing plutonium, iron, and aluminum. A method was developed for separating aluminum and iron prior to color development.

Excess EDTA (ethylenediaminetetraacetic acid) was added to the sample to complex the iron and aluminum. Then uranium was complexed with 8-quinolinol and extracted with chloroform. A back extraction with aqueous ammonium carbonate removed the uranium-234 and left plutonium and iron in the organic phase. Nitric acid was added, and the sample was heated to dryness to destroy the carbonate and the 8-quinolinol.

The residue was dissolved in dilute nitric acid. A buffer and potassium cyanide (to complex any copper impurities) were added, and the pH was adjusted to 10 with ammonium hydroxide.

The uranium was complexed with 1-(2-pyridylazo)-2-naphthol (PAN) and dissolved in o-dichlorobenzene. After one hour to develop the color, the absorbance was read at 570 millimicrons. Beer's law was followed between 0 and 80 micrograms of uranium.

When extraneous metallic ions were removed, the uranium was complexed and analyzed spectrophotometrically without interference. A uranium-EDTA complex, which would cause low results, was also avoided.

Standard uranium solutions were processed through the separation of iron and aluminum; no uranium was lost during these steps.

ISOTOPE SEPARATION

Processes are being developed for separating and purifying the isotopes of a number of elements including hydrogen, the noble gases, carbon and uranium. Potential sources of supply of these materials are being evaluated.

Carbon-13

Gram quantities of greater than 90 per cent purity carbon-13 are being prepared. Various thermal diffusion column configurations are being used for separation efficiency studies and evaluation of methane and carbon monoxide as feed gases.

The first five stages of the seven-stage cascade system of hot-wire and concentric-tube columns are enriching natural methane to about 23 per cent carbon-13 while coming to steady state conditions. The concentric tube columns have concentrated enriched methane from 60 per cent to over 90 per cent carbon-13. About 1.5 grams of product containing 10 per cent impurities have been accumulated.

Thermal Diffusion Column Wire Heating by Alternating Current

The use of alternating current for heating the center wires of thermal diffusion columns is being evaluated.[5] The effect of alignment of electrical leads on wire vibration is being investigated using direct current. To aid in wire alignment two parallel, flat iron discs two inches in diameter were used in the pool of mercury. The 1/16 inch Nichrome V column wire passed through the center of one disc and the battery connection through the other. The distance between the discs was approximately ¾ inch when the 36-inch wire was heated. There was no wire deflection when 48 amperes was applied at 24 volts, nor did the wire vibrate when alternating current was used. The discs, however, did not prevent vibration in a 12-foot, 1/16-inch diameter, Nichrome V wire when direct current was applied. Standing waves which slowly changed length formed in the wire. The mounting bracket at the top of the wire and the container of mercury on the floor were then shock-mounted using specially designed rubber pads. Shock mounting allowed the wire to have a true standing wave.

Next, the effect of the mercury pool on the vibrations was studied. The mercury pool was replaced by two rubber bands attached at an angle on the wire above the attached weight. An electrical connection was made above the weight directly to the Nichrome V wire. There was no vibration in the wire up to 25 amperes of alternating current. The weight was then removed, and the tension on the Nichrome V wire was imposed by the rubber bands only. No vibration of the wire occurred with heating. A spring attached in the same axis as the wire in place of the rubber bands resulted in slight wire vibration.

The electrical connection to the wire was evaluated for its vibration damping effect. A braided cable was used to conduct alternating current and also to act as a vibration damper in a horizontal plane. Both the vibration and the use of a mercury pool were eliminated completely. The braided cable expanded and contracted as the wire was heated or cooled, and the spring inside the cable maintained tension on the wire.

[5] MLM-1160.

ALPHA AND NEUTRON SOURCE DEVELOPMENT

Mound Laboratory is responsible for manufacturing alpha and neutron sources from polonium-210 and plutonium-239, which cannot be produced by American industry at the present time. The techniques of fabricating these unusual sources are being developed and publicized.

Neutron Measurements

An approximate integral was set up to determine the error introduced into the calibration of neutron sources due to the finite geometry of a source. A program was written to evaluate this integral by numerical methods. A check on the accuracy of this analysis is possible by comparing the flux distribution about a source, as predicted by the integral, with that actually measured. As the program is now written, there is about an eight per cent discrepancy between the predicted and measured distributions. A small error was discovered in the integral which, corrected, should reduce the discrepancy, though not enough to give perfect agreement. This lack of perfect agreement will not seriously affect the evaluation of the influence of source geometry on calibration. For a source with 80 grams of plutonium, the effect of geometry gave a calibration 0.25 per cent higher than for a point source.

The ratio of room-scattered neutrons to unscattered neutrons in the counting room is being determined. This ratio has now been obtained at four different source-to-detector separations for Po-Be and Pu-Be neutrons by using four shadow shields. An empirical expression has been obtained for the ratio as a function of source-to-detector separation; this expression is being used in an analysis of data from inverse-square runs.

Previously, data had been fitted to the following expression:

$$y = a(1 + d\,x) + \frac{b}{(x+c)^2}$$

where y is the measured counting rate, $a(1+dx)$ is the counting rate due to room-scattered neutrons and $b/(x+c)^2$ is the counting rate due to unscattered neutrons. The factor $(1+dx)$ expresses the variation in room scattering with the measured source-to-detector separation x; it had previously been determined from measurements with a single shadow shield. The factor b is proportional to the source emission rate in the direction of the detector, and $(x+c)$ is the separation between the source and the effective center of the detector (c is a function of neutron energy). Fitting data from an inverse square run gave the parameters a, b, and c.

A new form for the above equation was obtained in which the room-scattered component is completely determined by measurements with shadow shields, and only the parameters b and c are obtained in the adjustment of the counting data. This new equation is written as follows:

$$y = \frac{b}{(x+c)^2}[1+f(x)]$$

where $f(x)$ is the ratio of room-scattered neutrons to unscattered neutrons. Since only two parameters are now being adjusted, values of b and c will be more consistant than when three parameters were adjusted. A computer program is being written for this new inverse-square expression.

Polonium Alpha Sources

Several experimental plated alpha sources were prepared by different plating techniques. Copper was used as the base material because the polonium is more uniform than when it is plated on platinum. If the source is removed from the polonium bath, rinsed, and transferred wet to the subsequent baths, less solution of the polonium occurs and thinner wipe-free coverage is possible.

The use of a silver strike bath after polonium plating but before gold plating results in a thinner, wipe-free gold plating. Silver will dissolve preferentially to polonium in subsequent plating, and polonium which dissolves is immediately redeposited by electrochemical exchange with the silver.

In previous plating of sources, gold was deposited in a series of plating increments alternating with scouring of the source to swage down and seal the gold surface against migration of polonium during subsequent platings. Photomicrographs of the scoured surfaces showed that the powder size was so large that grooves were being cut nearly through the gold. Rouge was substituted, and it has given better results.

ANALYTICAL

Methods of analyzing elements and compounds are being developed to support other programs at Mound Laboratory. These methods include instrumental techniques and classical wet methods.

Low-level Krypton Counting

A proportional counting gas (P-10) of 90 per cent xenon (plus a trace amount of krypton-85) and 10 per cent methane was expanded into a Johnston one-liter counting chamber at 600 torr. The net counting rate of the sample inside the low-level lead shield was 31,884 cpm. The krypton-85 content of this gas was 1.34×10^{-9} atom per cent. Thus, the efficiency of this counting chamber for krypton-85 is 0.68, and the sensitivity is 2.9×10^{-13} atom per cent.

The background count of the low-level lead shield was measured before and after the krypton was counted. The counting chamber was filled with P-10 gas to 600 torr. Prior to each loading with P-10 gas, the gas handling system and Johnston chamber were flushed with nitrogen gas. The P-10 gas was maintained in the gas handling system at approximately two atmospheres for 12 hours prior to expansion into the Johnston chamber. The background counting rate one hour after the chamber was loaded with P-10 gas increased by a factor of 2.4. A second flushing of the gas handling system and Johnston chamber reduced the background count one hour after loading by a factor of 0.6. The latter measurement was still 1.4 times the background count rate obtained before krypton-85 was counted in the chamber. A third and fourth flushing with nitrogen gas did not reduce the count sifnificantly. The high background in the chamber may restrict its use to gas samples known to contain traces of krypton-85.

A low-level beta counter was installed next to the low-level krypton system for comparison of the systems. The two systems had essentially identical shielding properties.

Four Pi Beta Counting

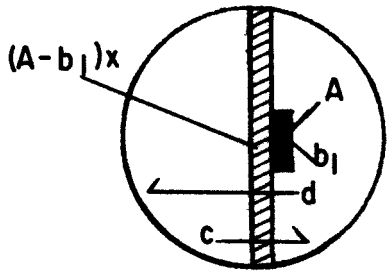
A study was made to verify that the counts in coincidence between the two hemispheres of the four pi beta counter were chiefly the result of cross scattering. This assumption of cross scattering is basic to the mathematics involved in absolute counting. Suzor and Charpak[6] attributed these coincidences to secondary electrons or photons produced in the sample mount in their experiments, an effect which was independent of the areas of the foil separating the two hemispheres.

An experiment was performed to show that secondary electrons did not produce the coincidence counts under the counting conditions at Mound Laboratory. Three sample holders were prepared from 0.005-inch lead foil, with apertures varying from 7/8 inch to 1¾ inches in diameter. A sample of Sr^{90}-Y^{90} mounted on 1/8 mil stainless steel attached to each of these holders was counted. Counts were made for all foil arrangements (Figure 8). The lead sample holders were sufficiently thick to absorb the strontium-90 betas; they were not entirely opaque to yttrium-90 betas. Thicker lead would have changed the geometry and introduced scattering from the lead.

Results listed in Table 3 show that the number of coincidences (c + d, e + f, and g) increased with aperture size. The increase was not strictly linear with aperture area due to such factors as counting statistics, instrument resolving time uncertainty, and penetration of the lead holder by scattered particles; however, the data substantiate the premise that the preponderant effect is cross scattering.

[6] F. Suzor and G. Charpak, "Investigation Concerning the Energy of Bachscattering of Electrons," *J. Phys. Radium*, 13, 1-10 (1952).

Case I. Single Foil

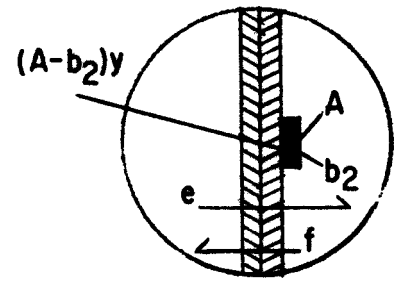


$R_1 = A + b_1 + d$

$L_1 = (A - b_1)x + c$

$S_1 = (A + b_1) + (A - b_1)x$

Case II. Two Foils

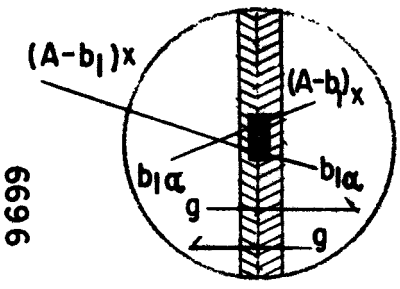


$R_2 = A + b_2 + f$

$L_2 = (A - b_2)y + e$

$S_1 = (A + b_2) + (A - b_2)y$

Case III. Two Foils (Sandwiched Source)



$R_3 = L_3 = (A - b_1)x + b_1\alpha + g$

$S_3 = 2[(A - b_1)x + b_1\alpha]$

9699

$c > e > d > g > f$

Figure 8. *Cross Section of Four Pi Counter.*

The values of all terms were computed from these data and are listed in Table 4. Terms c, e, d, g, and f increase with aperture size, while b_1, b_2, x, alpha, and 2A remain constant within the statistical errors of the measurements. This is evidence that no serious error results from the assumption that all of the coincidences, after resolving time corrections, are due to cross scattered particles.

Table 3

VARIATION OF COINCIDENCE COUNTS WITH APERTURE SIZE

Diameter of Aperture	Counts per Minute			
(Case I)	R_1	L_1	S_1	c + d
1¾	84,296	58,671	133,192	9774
1⅝	83,392	56,194	133,080	6506
⅞	82,967	54,524	133,805	3685
(Case II)	R_2	L_2	S_2	e + f
1¾	88,433	51,234	132,173	7494
1⅝	86,834	50,050	131,553	5330
⅞	86,825	48,944	131,963	3305
(Case III)	$R_3 = L_3$		S_3	2g
1¾	66,105		125,269	6940
1⅝	65,085		124,977	5192
⅞	63,366		124,121	2610

Table 4

VARIATION OF COMPUTED VALUES WITH APERTURE SIZE

Aperture Size (in.)	c*	e*	d*	g*	f*	b_1*	b_2*	x*	γ*	2A**
1¾	5566	5441	4208	3470	2052	12,813	19,002	0.975	0.744	134,576
1⅝	3606	3530	2900	2596	1800	13,002	17,548	0.965	0.762	134,980
⅞	2122	2020	1563	1305	1285	13,390	17,026	0.959	0.721	136,028

* Counts per minute

** Disintegrations per minute.

The nomenclature for Figure 8, a schematic of the cross section of the four pi chamber for each sample and foil combination discussed above, is as follows:

A = The number of particles emitted per unit time in the direction of the right hemisphere. (2A = 4 times disintegration rate of sample)

b_1 and b_2 = the number of particles emitted per unit time toward the left, but backscattered by the sample foil into the right hemisphere in Cases I and II.

c, d, e, f and g = the number of particles per unit time cross scattered into and counted in one hemisphere after having first been counted in the other.

x = absorption correction for a single foil for forward radiation (ratio of number of particles emerging from the foil to the number which strike the foil).

y = absorption correction for double foil for forward radiation.

α = absorption correction for a single foil for backscattered radiation.

R_1, L_1, S_1, R_2, L_2, S_2, R_3, L_3, S_3 are the measured counting rates in the right and left hemispheres and in both hemispheres in anticoincidence in Cases I, II, and III, respectively.